Probing Bose-Einstein Condensation of Excitons with Electromagnetic Radiation

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We examine the absorption spectrum of electromagnetic radiation from excitons, where an exciton in the $1s$ state absorbs a photon and makes a transition to the $2p$ state. We demonstrate that the absorption spectrum depends strongly on the quantum degeneracy of the exciton gas, and that it will generally manifest many-body effects. Based on our results we propose that absorption of infrared radiation could resolve recent contradictory experimental results on excitons in Cu_2O.

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The phenomenon of Bose-Einstein condensation has attracted much attention in the recent years, and many experimental groups have reported the formation of Bose-Einstein condensates in vapors of alkali-metal atoms [1]. Excitons, bound states of electrons and holes in semiconducting materials, are another candidate for undergoing this phase transition [2]. Since excitons are composite particles consisting of two fermions, they are expected to obey Bose-Einstein statistics in the limit where their thermal de Broglie wavelength becomes comparable to their interparticle spacing, provided that this spacing is much larger than the exciton Bohr radius.

Excitons in Cu_2O, which is a dipole forbidden material, have some important advantages in this respect [3]. Their radiative lifetime is long and their binding energy is much larger than the typical thermal energies. In addition the effective electron and hole masses are isotropic, and finally there are no bound states between excitons, i.e., biexcitons, or an electron-hole-liquid phase.

A lot of effort has been made in order to create a Bose-Einstein condensate of excitons in Cu_2O [4,5]. To determine the density and the temperature in the above experiments, the phonon-assisted recombination spectrum was fitted to a Bose-Einstein distribution, which gave the chemical potential and the temperature – two essentially independent parameters. Given the total exciton mass, the density was then evaluated to be on the order of 10^{18} cm^{-3}, while the temperature was on the order of $20-30$ K, higher than the lattice temperature which was kept at about 5 K. Therefore, according to this approach, the exciton gas was very close to the phase boundary for Bose-Einstein condensation, and the angular-momentum singlet-state (para)excitons were the species reported to actually cross the phase boundary.

More recent experiments [6] have, however, questioned the older method of determining the density and the temperature. In these experiments the number of excitons was determined directly and, with a relatively reliable estimate of the volume of the exciton cloud, the density was found to be two to three orders of magnitude lower, i.e., around 10^{16} cm^{-3}, where the exciton gas should show no sign of quantum degeneracy.

One, therefore, needs to find a reliable method of determining the density and in particular the degree of quantum degeneracy of excitons. In this study we propose that measuring the absorption spectrum of infrared radiation which induces transitions of the excitons from the $1s$ to the $2p$ state can indeed resolve the discrepancy. Our study is also directly applicable to other systems, like excitons in quantum wells, and thus it could help resolve some other experimental observations which are controversial [7].

Öktel and Levitov [8] have examined a similar problem as the one we consider here, in the context of optical excitations of hydrogen atoms and have studied the many-body effects that show up in the absorption spectrum, for an effective contact potential between the atoms. Our approach is equivalent to theirs in the limit of equal masses between the excitons in the $1s$ and the $2p$ state. In another study Pethick and Stoof [9] have considered a more general form of the interatomic potential. Finally Lewenstein and You [10] have examined the enhancement of scattering of light from a gas of bosonic atoms as they form a Bose-Einstein condensate, and have suggested that this effect could be used for the detection of the condensate.

This Letter is organized as follows: We first consider the case of an ideal exciton gas, and examine the relevant energy scales that enter the problem and also derive simple expressions for the absorption spectrum. We then examine the problem of an interacting exciton gas within the Hartree-Fock approximation and find that the interactions can have a very drastic effect on the absorption spectrum. Finally we present our results with the interactions included, and show that infrared absorption can be used in order to determine the degree of quantum degeneracy of excitons, thus proposing an experiment which could resolve this issue.

Consider the process in which an exciton in the $1s$ state, with momentum $\hbar\mathbf{k}$, absorbs a photon of momentum $\hbar\mathbf{q}$, making a transition to the $2p$ state with momentum $\hbar(\mathbf{k}+\mathbf{q})$. The conservation of energy in this process implies that

$$\epsilon^{1s}_{\mathbf{k}} + \hbar c q = \epsilon^{2p}_{\mathbf{k}+\mathbf{q}}, \tag{1}$$

where $\epsilon^{i}_{\mathbf{k}} = E_i + \hbar^2 k^2/2m_i$ with E_i being the binding energy of the i state, and m_i being the total exciton

mass in the state i. In Cu_2O $m_{1s} \approx 3m$, where m is the electron mass, is larger than the sum of the effective electron and hole masses as a result of the small Bohr radius of the $1s$ excitons, $a_B^{1s} \approx 5.3$ Å compared to the lattice constant $a_l \approx 4.26$ Å [11]. On the other hand, the Bohr radius of excitons in the $2p$ state a_B^{2p} is given by the hydrogenic formula which yields ≈ 44 Å [11]. Since $a_B^{2p} \gg a_l$, m_{2p} is expected to be equal to the sum of the effective electron and hole masses, which is $\approx 1.68m$. In Eq. (1) there are two distinct energy scales, i.e., the energy separation $\Delta E = E_{2p} - E_{1s} \approx 55$ meV, and the thermal energy $\hbar^2 k^2/2m \sim k_B T$, which is of order 1–10 meV. Since $\hbar qc \sim \Delta E$ and $\hbar^2 k^2/2m \sim k_B T$, we get that $q/k \sim \Delta E/\sqrt{mc^2 k_B T} \sim 10^{-3}$. Therefore $\hbar^2 qk/2m \sim 10^{-3} k_B T$, and $\hbar^2 q^2/2m \sim 10^{-6} k_B T$, which allows us to neglect the corresponding terms in Eq. (1). Solving in terms of $k^2(q)$, we obtain

$$k^2(q) \approx \frac{2m_{1s}m_{2p}}{m_{1s} - m_{2p}} \frac{\hbar cq - \Delta E}{\hbar^2}, \tag{2}$$

which gives the magnitude of the momentum $\hbar\mathbf{k}$ of the exciton in the $1s$ state that absorbs a photon with wavevector $\mathbf{q}$ and is excited to the $2p$ state. In this approximation, for $\hbar cq = \Delta E$ only the excitons with $k = 0$ can participate in the process; however for a Bose-Einstein condensate there is a macroscopic number of excitons with $k = 0$, and therefore the absorption spectrum has a pronounced peak, with a strong temperature dependence. To see this more clearly, let us calculate the rate of this dipole-allowed process of absorption of a photon. With the approximate expression for the conservation of energy of Eq. (2), the rate Γ_T of non-condensed excitons in the $1s$ state absorbing a photon and making the transition to the $2p$ state is given by

$$\Gamma_T = \frac{2\pi}{\hbar} \sum_{\mathbf{k}} |M_{\mathbf{q}}|^2 n_{\mathbf{k}}^{1s} (1 + n_{\mathbf{k}+\mathbf{q}}^{2p}) f_{\mathbf{q}}\, \delta(\hbar qc - \Delta E_{\mathbf{k}}), \tag{3}$$

where $M_{\mathbf{q}}$ is the matrix element of this process, $n_{\mathbf{k}}^i$ is the distribution function of species i ($1s$ or $2p$ excitons), $f_{\mathbf{q}}$ is the distribution function of the incoming photons, and $\Delta E_{\mathbf{k}} = \epsilon_{\mathbf{k}}^{2p} - \epsilon_{\mathbf{k}}^{1s}$. Neglecting the occupation number of excitons in the $2p$ state, $n_{\mathbf{k}+\mathbf{q}}^{2p} \ll 1$, for monochromatic radiation Eq. (3) implies that

$$\Gamma_T \propto (\hbar cq - \Delta E)^{1/2} n_{\mathbf{k}_0}^{1s}\, \theta(\hbar cq - \Delta E), \tag{4}$$

where $\theta(x)$ is the Heaviside step function, and the magnitude of $\mathbf{k}_0$ is given by Eq. (2). The above result expresses the fact that the absorption spectrum is proportional to the density of states times the distribution function calculated at a wavevector with a magnitude given by Eq. (2).

For a Bose-Einstein condensed exciton gas with N_C excitons occupying the $\mathbf{k} = 0$ state, the rate Γ_C of the same process is simply

$$\Gamma_C = \frac{2\pi}{\hbar} |M_{\mathbf{q}}|^2 N_C (1 + n_{\mathbf{q}}^{2p}) f_{\mathbf{q}}\, \delta(\hbar qc - \Delta E), \tag{5}$$

or $\Gamma_C \propto N_C\, \delta(\hbar cq - \Delta E)$. Therefore the absorption spectrum (which is proportional to the decay rate) of an ideal Bose-Einstein condensed gas has a strong peak with a height that scales as N_C. However, as we show below, the interactions can modify this picture drastically.

We thus turn to the more realistic problem of an interacting Bose gas. We start with the Hamiltonian H [12]

$$H = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}}^{1s} a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \frac{U_{11}}{2V} \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} a_{\mathbf{k}+\mathbf{q}}^\dagger a_{\mathbf{k}'-\mathbf{q}}^\dagger a_{\mathbf{k}'} a_{\mathbf{k}} + \sum_{\mathbf{k}} \epsilon_{\mathbf{k}}^{2p} b_{\mathbf{k}}^\dagger b_{\mathbf{k}} + \frac{U_{12}}{V} \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} b_{\mathbf{k}+\mathbf{q}}^\dagger a_{\mathbf{k}'-\mathbf{q}}^\dagger a_{\mathbf{k}'} b_{\mathbf{k}}, \tag{6}$$

where V is the volume of the gas. In the above Hamiltonian we assume that the excitons interact with an effective contact potential, with $U_{ij} = 2\pi\hbar^2 a_{ij}/\mu_{ij}$ being the strength of the effective two-body interaction. Here a_{ij} is the scattering length for collisions between excitons in the states i and j (1 for the $1s$ state, and 2 for the $2p$ state). The reduced mass μ_{ij} entering the above expression is given by $\mu_{ij} = m_i m_j/(m_i + m_j)$. Also $a_{\mathbf{k}}(b_{\mathbf{k}})$ and $a_{\mathbf{k}}^\dagger(b_{\mathbf{k}}^\dagger)$ are annihilation and creation operators for an exciton with momentum $\mathbf{k}$ in the $1s(2p)$ state.

Let us now consider the ground state of the system with N excitons, which we denote as $|0\rangle = |N_C, N_{\mathbf{k}_1}, N_{\mathbf{k}_2}, \ldots, N_{\mathbf{k}_e}, \ldots\rangle$, where $N_{\mathbf{k}_i}$ is the occupancy of a state with momentum $\mathbf{k}_i$. Initially we take all the excitons to be in the $1s$ state. Since we consider a Bose gas both in the normal, as well as in the condensed regime, we assume that there is one state that can get populated by a macroscopic number of excitons, N_C, and thus N_C can get as high as the total number of excitons, N, whereas the $N_{\mathbf{k}_i}$ are of order unity.

We now examine such a system when one creates excitations of the excitons from the $1s$ to the $2p$ state with the action of some laser pulse. If an exciton with momentum $\mathbf{k}_e$ is excited to the $2p$ state with momentum $\mathbf{k}_e' = \mathbf{k}_e + \mathbf{q}$, where $\mathbf{q}$ is the wavevector of the laser light, since $\mathbf{q}$ is very small, we shall assume that we have vertical transitions, i.e., $\mathbf{q} = 0$. We denote the excited states as $|\Phi_{\text{exc},\mathbf{k}_e}\rangle = |\mathbf{k}_e; N_C, N_{\mathbf{k}_1}, N_{\mathbf{k}_2}, \ldots, N_{\mathbf{k}_e} - 1, \ldots\rangle$, which are the basis vectors of our problem. The number of such states is $N - N_C + 1 = N_T + 1$, where $N_T = N - N_C$ is the number of $1s$ excitons in states with $\mathbf{k} \neq 0$. The laser beam that excites the excitons from the $1s$ to the $2p$ state creates a superposition of the states $|\Phi_{\text{exc},\mathbf{k}_e}\rangle$ [12]. Thus, in order to determine the absorption spectrum, we consider the matrix with elements $H_{i,j} = \langle\Phi_{\text{exc},\mathbf{k}_i}|H|\Phi_{\text{exc},\mathbf{k}_j}\rangle - \langle 0|H|0\rangle$. One finds that

$$H_{i,j} = \delta_{i,j}[\Delta E_{\mathbf{k}_i} + U_{11}(n_{\mathbf{k}_i} - 2n) + U_{12}(n - n_{\mathbf{k}_i})] + U_{12}\sqrt{n_{\mathbf{k}_i} n_{\mathbf{k}_j}}, \tag{7}$$

where $n = N/V$ and $n_{\mathbf{k}_i} = n_{\mathbf{k}_i}^{1s} = N_{\mathbf{k}_i}/V$ is the Bose-Einstein distribution for the $1s$ excitons. Let Ψ_i be the

components of an eigenvector with eigenvalue E. Starting from the eigenvalue equation $\sum_{j=0}^{N_T} H_{i,j}\Psi_j = E\Psi_i$, we solve in terms of Ψ_i, multiply by $\sqrt{n_{\mathbf{k}_i}}$ and sum over i. Eliminating the factor $\sum_{j=0}^{N_T}\Psi_j\sqrt{n_{\mathbf{k}_j}}$ from the resulting equation, the eigenvalues of $H_{i,j}$ are then given by the roots of $g(E)-1=0$, where

$$g(E) = \sum_{i=0}^{N_T} \frac{U_{12} n_{\mathbf{k}_i}}{E - [\Delta E_{\mathbf{k}_i} + U_{11}(n_{\mathbf{k}_i} - 2n) + U_{12}(n - n_{\mathbf{k}_i})]}. \tag{8}$$

Distinguishing the condensed state ($i=0$) from the other states ($i \neq 0$), Eq. (8) takes the following form in the thermodynamic limit

$$g(E) = \frac{U_{12} n_c}{E - [\Delta E + U_{11}(n_c - 2n) + U_{12}(n - n_c)]} + \sum_{i\neq 0}^{N_T} \frac{U_{12} n_{\mathbf{k}_i}}{E - [\Delta E_{\mathbf{k}_i} + n(U_{12} - 2U_{11})]}, \tag{9}$$

where $n_C = N_C/N$. In the above equation there are in general three limiting cases, depending on the ratio of the interaction energy nU_{12}, to the typical kinetic energy $\Delta E_{\mathbf{k}_i}$, which is on the order of the thermal energy, k_BT. In the limit $nU_{12} \ll k_BT$, one recovers the results we found earlier for the ideal Bose gas. In the opposite limit, $nU_{12} \gg k_BT$, the behaviour of the system of excitons is “collective”. A graphical solution of the eigenvalue equation shows that in the condensed phase, where both N_C and N_T are of order N, there are two strong modes, which give rise to two peaks in the absorption spectrum. There are also $N_T - 1$ solutions, which form a continuum corresponding to single-particle excitations of the thermal excitons. In the same limit $nU_{12} \gg k_BT$ for a fully condensed gas as well as for a gas in the normal state, there is only one mode, since then one has a one-component system. Finally when $nU_{12} \sim k_BT$ the system behaves in a “mixed” way. In addition, the limit $|m_{1s}/m_{2p} - 1| \ll 1$, is equivalent to the case $nU_{12} \gg k_BT$ and Eq. (9) reduces to a quadratic algebraic equation, which gives the same result as the one derived by Öktel and Levitov in Ref. [8].

By adding a small imaginary part in $g(E)$, i.e., $g(E + i\eta)$, where finite η results in homogeneous broadening of the energy levels, we calculate the corresponding imaginary part of the susceptibility $[g(E+i\eta)-1]^{-1}$ obtaining the absorption spectra shown in Fig. 1. Broadening can be calculated from first principles [9] – however in the present study we assume small homogeneous broadening, choosing $\eta = 10^{-2}$ meV to produce the graphs in Fig. 1. The broadening of the energy levels is expected to be small, and this can be seen by examining the three basic mechanisms which contribute to that, i.e., the exciton-exciton elastic collisions, their scattering with the lattice, and their radiative lifetime. The radiative lifetime of the orthoexcitons in the $1s$ state τ_o^{1s} is $\sim 10^{-5}$s [6] and that of the paraexcitons τ_p^{1s} is $\sim 10^{-3}$s. The radiative lifetime in the $2p$ state τ_i^{2p} is smaller by a factor of $(k_\gamma a_B^{2p})^2$, since the transition is dipole allowed, where k_γ is the wavevector of the emitted photon. Since $k_\gamma = E_g/\hbar c$, where $E_g \approx 2.17$ eV is the gap energy, $k_\gamma \approx 10^{-3}$ Å^{-1}. Therefore $\tau_o^{2p} \sim 10^{-8}$ s, while $\tau_p^{2p} \sim 10^{-6}$ s. The exciton-phonon scattering time is on the order of 10^{-9}s [13]. Finally for a density as high as 10^{18} cm^{-3}, the exciton-exciton scattering time is on the order of 10^{-11}s [14], which turns out to be the shortest possible timescale, giving an energy broadening of less than 0.1 meV.

We now analyze the results shown in Fig. 1. To produce these graphs, we made use of the results of Ref. [15], that $a_{11} = 2.1 a_B^{1s}$ for paraexcitons, and assumed that $a_{11} = 10$ Å. For the value of a_{12} very little is known and for this reason we have considered both the case of positive (left column in Fig. 1), as well as negative (right column in Fig. 1) values. The ratio $|a_{12}/a_{11}|$ is expected to be larger than 1, since $a_B^{2p} = 44$ Å, which is much larger than $a_B^{1s} \approx 5$ Å. We made the conservative choice $|a_{12}/a_{11}| = 2$, although this ratio could be larger. We also considered a temperature of 10 K for the exciton gas in all the cases, and we varied the density from 10^{16} cm^{-3} to 5×10^{18} cm^{-3}. With these values $k_BT \sim 1$ meV, while $|nU_{12}|$ is $\sim 10^{-2}$ meV for $n = 10^{16}$ cm^{-3}, and ~ 5 meV for $n = 5\times 10^{18}$ cm^{-3}. Figures 1(a+) and (a–) show a completely classical gas, and since $nU_{12} \ll k_BT$, the system behaves like an ideal gas. In Figs. 1(b+) and (b–) the excitons are essentially on the phase boundary for condensation and since $nU_{12} \sim k_BT$, the system is in the “mixed” state where both collective and single-particle-like behaviours show up. Figure 1(b–) shows these two distinct types of excitation, while Fig. 1(b+) does not, because the collective mode is buried inside the continuum. In Figs. 1(c+) and (c–) the excitons are in the condensed phase with $N_C/N \approx 0.48$. This is the source of the sudden appearance of the peak in Fig. 1(c+). In Fig. 1(c–) in addition to the two peaks, there is a contribution from the continuum that is hardly visible. Finally in Figs. 1(d+) and (d–) $N_C/N \approx 0.79$, and since nU_{12} is about $5k_BT$, the spectrum is dominated by the collective behaviour, as the two peaks indicate. However, we remark that the Hartree-Fock approximation does not capture effects due to condensate fluctuations which may be relevant in the regime $nU_{12} \gg k_BT$.

Let us now examine the possible experiment which could be performed in order for these effects to be explored. The energy of the absorbed radiation would have to be in the infrared, with an energy of order $\Delta E \approx 55$ meV. The corresponding wavelength is about 20 μm, and it is comparable to the size of the cloud. Free-electron lasers provide tunable radiation in this regime. It is important to mention that at such low energies the crystal is transparent and the absorption of radiation due to the process we study should be the dominant mechanism. Our analysis requires that the infrared pulse should be sufficiently long, so that its energy spread is much less

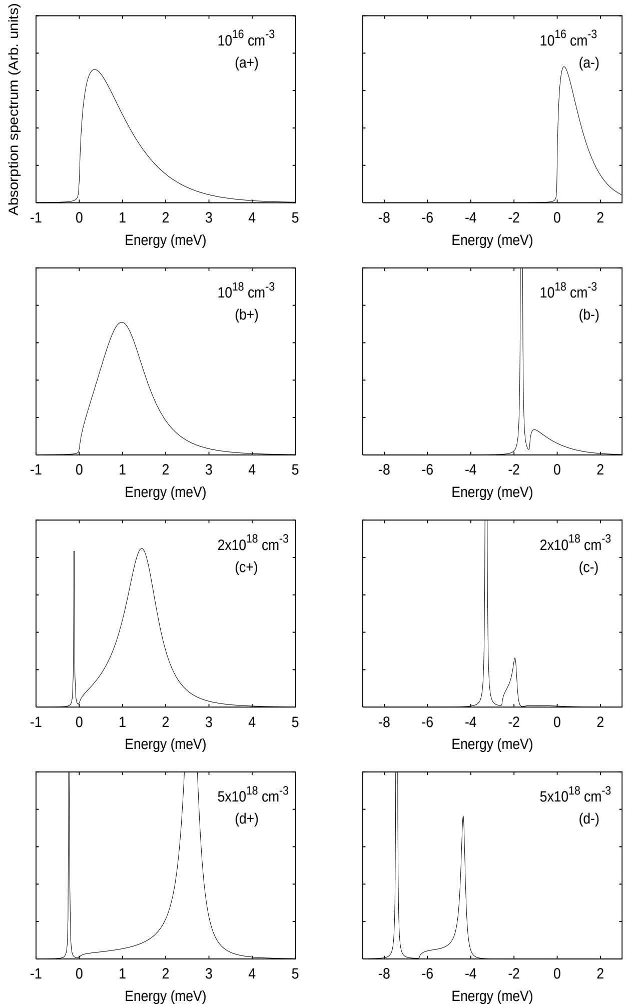

FIG. 1. The absorption spectrum on a linear scale, as a function of the energy of the photon that is absorbed. The absorption is in arbitrary units, but the scale is the same in all the figures. The energy is measured in units of meV, and its zero is measured with respect to ΔE. The scattering length $a_{11} = 10$ Å, the exciton temperature is 10 K, and the density is 10^{16}, 10^{18}, 2×10^{18}, and 5×10^{18} cm^{-3} from top to bottom. For the graphs on the left $a_{12} = 20$ Å, and for the ones on the right, $a_{12} = -20$ Å. If μ is the chemical potential of the gas, $-\mu/k_B T = 3.7$ in (a$\pm$), and 5×10^{-4} in (b$\pm$), while $N_C/N = 0.48$ in (c$\pm$), and 0.79 in (d$\pm$).

than the energy width of the structures shown in Fig. 1. An advantage of the method we suggest is that it provides an independent method of probing the kinetic energy distribution of excitons. The difference between the uppermost graphs in Fig. 1 and the lowest is pronounced, and one should be able to distinguish clearly the degree of degeneracy of the excitons. In addition, this method does not depend on the strength of the phonon-assisted recombination line of paraexcitons, which is very weak, and since it is close to other much stronger lines, observing this line is very hard [4–6].

In conclusion, we have demonstrated that the absorption spectrum of electromagnetic radiation which induces transitions of the excitons from the $1s$ to the $2p$ state is strongly affected by the quantum degeneracy of the exciton gas and by many-body effects. We have thus concluded that the absorption spectrum can provide a powerful tool for resolving recent experimental contradictory results on excitons in Cu_2O, and in quantum wells.

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